Self-diffusion in sheared colloidal suspensions: violation of fluctuation-dissipation relation

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Using memory-function formalism we show that in sheared colloidal suspensions the fluctuation-dissipation theorem for self-diffusion, *i.e* Einstein's relation between self-diffusion and mobility tensors, is violated and propose a new way to measure this violation in Brownian Dynamics simulations. We derive mode-coupling expressions for the tagged particle friction tensor and for an effective, shear-rate dependent temperature.

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There has been a lot of interest recently in non-equilibrium behavior of colloidal systems [1]. On the practical side, it has been stimulated by the importance of non-equilibrium properties for the preparation and processing of colloidal materials. From the more fundamental perspective, colloidal suspensions serve as model soft-glassy systems: they have properties similar to those of more complex soft materials but are simple enough to allow for detailed microscopic, experimental and theoretical investigations. Additional impetus came from analysis of simple statistical mechanical models (*i.e.* fully connected spin systems) that predicted violation of the fluctuation-dissipation theorem (FDT) out of equilibrium [2], and an intriguing connection between non-equilibrium and glassy properties [3]. Subsequently, FDT violation was found by means of computer simulations in model supercooled fluids [4, 5].

Although non-equilibrium phenomenology of colloidal suspensions is reasonably well described by so-called "schematic models" [6, 7], it is of great fundamental and practical interest to develop more microscopic approaches. First, connection between non-equilibrium and glassy behavior, if it also exists for colloidal systems, may provide new insights into the glass transition problem. Second, simulational and experimental studies of colloidal systems provide detailed information that cannot be described in terms of schematic models. Third, a more microscopic approach would allow one to correlate microscopic properties and macroscopic behavior.

Recent investigations of the non-equilibrium behavior can be divided into two categories. Initially, transient behavior of glassy systems, *i.e.* aging, attracted the most attention [2, 3, 4]. Microscopic, theoretical analysis of aging is in its infancy [8]. More recently, relaxation under steady shear was investigated [5, 6, 9]. Analysis of sheared suspensions is easier: stationary nature of the shear flow restores time-translational invariance and thus simplifies the problem both conceptually and technically.

Colloidal suspensions under steady shear flow were the subject of two recent theoretical investigations [10, 11]. Both approaches were based on the "least inadequate" [12] microscopic theory of colloidal dynamics: mode-coupling theory (MCT) [13]. The difference between them parallels the difference between derivations of MCT for non-sheared colloids: Ref. [10] used the projection operator method whereas Ref. [11] started from generalized fluctuating hydrodynamics. Both approaches recovered the most important features of the soft-glassy rheology: accelerated relaxation of sheared colloidal fluids and shear melting of the colloidal glass. These phenomena were attributed to flow-induced advection of density fluctuations and the resulting perturbation of the "cage effect". Neither work, however, addressed FDT violation.

The goal of this Letter is to investigate the origin of FDT violation for the simplest possible process: self-diffusion (*i.e.* diffusion of a tagged particle) in a sheared colloidal suspension [14, 15]. In this case FDT amounts to Einstein's relation between the self-diffusion tensor and the tagged particle mobility tensor. We generalize the conventional memory function description of self-diffusion to colloidal systems under shear and derive a Green-Kubo-like relation for the self-diffusion tensor in a sheared suspension. Next, we derive a Green-Kubo-like relation for the tagged particle mobility tensor. We show that FDT violation is associated with the non-equilibrium nature of the stationary, shear-rate dependent probability distribution. We propose a new approach to monitor FDT violation in Brownian Dynamics simulations that does not require introducing an external perturbation. Finally, we use the memory function approach to derive MCT expressions for the tagged particle friction tensor and an effective temperature.

We start with the definition of the self-intermediate scattering function, $F_s(\mathbf{k}_1;\mathbf{k}_2;t)$:

$$F_s(\mathbf{k}_1;\mathbf{k}_2;t) = \left< n_s(\mathbf{k}_1)\exp(\Omega t) n_s(-\mathbf{k}_2)\right> . \qquad (1)$$

Here $n_s(\mathbf{k}_1)$ is the Fourier transform of the microscopic density of particle number 1, *i.e.* the tagged particle, $n_s(\mathbf{k}_1) = e^{-i\mathbf{k}_1\cdot\mathbf{r}_1}$. Furthermore, Ω is the N-particle evolution operator, *i.e.* the Smoluchowski operator [16],

$$\Omega = -D_0 \sum_l \frac{\partial}{\partial \mathbf{r}_l}\cdot\left(-\frac{\partial}{\partial \mathbf{r}_l} + \beta\mathbf{F}_l + \mathbf{v}(\mathbf{r}_l)\right), \qquad (2)$$

where D_0 is the diffusion coefficient of an isolated colloidal particle, $\beta = 1/(k_B T)$, $\mathbf{F}_l$ is a force acting on the lth particle, and $\mathbf{v}(\mathbf{r}) = \mathbf{\Gamma} \cdot \mathbf{r}$ is the shear flow with $\mathbf{\Gamma} = \dot{\gamma}\hat{\mathbf{x}}\hat{\mathbf{y}}$ being the velocity gradient tensor ($\dot{\gamma}$ denotes the shear rate). Finally, in Eq. (1) $\langle \ldots \rangle$ denotes the stationary, shear-rate dependent ensemble average at temperature T. The probability distribution stands to the right of the quantity being averaged and all operators act on it as well as on everything else. Note that translational symmetry leads to $F_s(\mathbf{k}_1;\mathbf{k}_2;t) \propto \delta_{\mathbf{k}_1(t),\mathbf{k}_2}$ where $\mathbf{k}_1(t) = \mathbf{k}_1 + {}^t\mathbf{\Gamma} \cdot \mathbf{k}_1 t$ with ${}^t\mathbf{\Gamma}$ being the transpose of $\mathbf{\Gamma}$.

To derive the memory function representation we start from the exact expression for the Laplace transform (LT) of the time derivative of $F_s(\mathbf{k}_1;\mathbf{k}_2;t)$,

$$LT(\dot{F}_s) = \left\langle n_s(\mathbf{k}_1)\Omega \frac{1}{z-\Omega} n_s(-\mathbf{k}_2) \right\rangle. \tag{3}$$

We define the projection operator on the space spanned by the tagged particle density,

$$\hat{P}_s \;=\; \sum_{\mathbf{q}} \ldots n_s(-\mathbf{q}) \,\rangle \langle\, n_s(\mathbf{q}) \ldots . \tag{4}$$

Note that, in contrast to Ref. [10], the definition of $\hat{P}$ involves the stationary, shear-rate dependent distribution. Next, we follow the usual projection operator manipulations and arrive at the following memory function representation of the time derivative of $F_s(\mathbf{k}_1;\mathbf{k}_2;t)$,

$$\begin{aligned} LT(\dot{F}_s) \;=\; & \sum_{\mathbf{k}_3} \langle n_s(\mathbf{k}_1)\Omega n_s(\mathbf{k}_3)\rangle \, F_s(\mathbf{k}_3,\mathbf{k}_2;z) \\ & + \sum_{\mathbf{k}_3} \left\langle n_s(\mathbf{k}_1)\Omega \hat{Q}_s \frac{1}{z-\hat{Q}_s\Omega\hat{Q}_s} \hat{Q}_s \Omega n_s(\mathbf{k}_3) \right\rangle \\ & \times F_s(\mathbf{k}_3,\mathbf{k}_2;z), \end{aligned} \tag{5}$$

where $\hat{Q}_s$ is the projection on the subspace orthogonal to the tagged particle density, $\hat{Q}_s = 1 - \hat{P}_s$.

Using translational invariance of the sheared suspension and the fact that in the stationary state the average force acting on the tagged particle vanishes, we get

$$\langle n_s(\mathbf{k}_1)\Omega n_s(\mathbf{k}_3)\rangle = -D_0 k_1^2 \delta_{\mathbf{k}_1,\mathbf{k}_3} + \mathbf{k}_1 \cdot \mathbf{\Gamma} \cdot \frac{\partial}{\partial \mathbf{k}_1} \delta_{\mathbf{k}_1,\mathbf{k}_3}, \tag{6}$$

where the second term on the right-hand-side (RHS) of Eq. (6) describes flow-induced advection.

Furthermore, using the explicit form of the evolution operator we obtain the following identity:

$$\begin{aligned} & \left\langle n_s(\mathbf{k}_1)\Omega \hat{Q}_s \frac{1}{z-\hat{Q}_s\Omega\hat{Q}_s} \hat{Q}_s \Omega n_s(\mathbf{k}_3) \right\rangle \\ & = \mathbf{k}_1 \cdot \left\langle \mathbf{j}_s(\mathbf{k}_1) \frac{1}{z-\hat{Q}_s\Omega\hat{Q}_s} \left(2\mathbf{j}_s^{\text{eff}}(-\mathbf{k}_3) - \mathbf{j}_s(-\mathbf{k}_3)\right) \right\rangle \cdot \mathbf{k}_3. \end{aligned} \tag{7}$$

Here $\mathbf{j}_s(\mathbf{k})$ is a projected tagged particle current density,

$$\mathbf{j}_s(\mathbf{k}) = \hat{Q}_s D_0 \left(-i\mathbf{k} + \beta\mathbf{F}_1 + \mathbf{v}_1\right) e^{-i\mathbf{k}\cdot\mathbf{r}_1}, \tag{8}$$

and $\mathbf{j}_s^{\text{eff}}(\mathbf{k})$ is a projected, effective current density,

$$\mathbf{j}_s^{\text{eff}}(\mathbf{k}) = \hat{Q}_s D_0 \left(-i\mathbf{k} + \beta\mathbf{F}_1^{\text{eff}}\right) e^{-i\mathbf{k}\cdot\mathbf{r}_1}. \tag{9}$$

In Eq. (9) $\mathbf{F}_1^{\text{eff}}$ is the effective force acting on the tagged particle that is defined in terms of the stationary, shear-rate dependent probability distribution,

$$\beta\mathbf{F}_1^{\text{eff}} = \frac{\partial}{\partial \mathbf{r}_1} \ln P^{\text{st}}(\mathbf{r}_1,\ldots,\mathbf{r}_N). \tag{10}$$

Combining Eqs. (6-10), in the small wavevector, long time limit we get

$$LT(\dot{F}_s) = \left(-\mathbf{k}_1 \cdot \mathbf{D} \cdot \mathbf{k}_1 + \mathbf{k}_1 \cdot \mathbf{\Gamma} \cdot \frac{\partial}{\partial \mathbf{k}_1}\right) F_s(\mathbf{k}_1,\mathbf{k}_2;z), \tag{11}$$

where the self-diffusion tensor $\mathbf{D}$ is given by the following Green-Kubo-like expression:

$$\mathbf{D} = D_0 - (\beta D_0)^2 \int_0^\infty dt \, \left\langle \mathbf{F}_1 \exp(\Omega t) \left(2\mathbf{F}_1^{\text{eff}} - \mathbf{F}_1\right) \right\rangle. \tag{12}$$

Note that in $\mathbf{k} \to 0$ limit projected dynamics (*i.e.* $\hat{Q}_s\Omega\hat{Q}_s$) can be replaced by real dynamics (*i.e.* Ω) [17].

To get the tagged particle mobility tensor we follow approach used by Lekkerkerker and Dhont [18]. A calculation along the lines of Sec. III of Ref. [18] leads to the following Green-Kubo-like formula for the long-time tagged particle mobility tensor μ (here $\mu_0 = \beta D_0$ is the mobility of an isolated colloidal particle):

$$\mu = \mu_0 - \mu_0^2 \beta \int_0^\infty dt \, \left\langle \mathbf{F}_1 \exp(\Omega t) \mathbf{F}_1^{\text{eff}} \right\rangle. \tag{13}$$

Comparison of Eqs. (12) and (13) shows that Einstein's relation between self-diffusion and mobility tensors is violated. The origin of the violation is the difference between the force acting on the tagged particle, $\mathbf{F}_1$, and the effective force, $\mathbf{F}_1^{\text{eff}}$. In the absence of the shear flow, $\mathbf{F}_1 = \mathbf{F}_1^{\text{eff}}$ and the usual Einstein relation follows. In the presence of the flow, one can follow Ref. [5] and use the transverse components of the self-diffusion and mobility tensors to define an effective, shear-rate dependent temperature T^{eff}, where $k_B T^{\text{eff}} = D_{zz}/\mu_{zz}$. Since we do not have an explicit expression for the effective force $\mathbf{F}_1^{\text{eff}}$, neither D_{zz} nor μ_{zz} can be obtained from direct simulational evaluation of its respective Green-Kubo-like expression. However, if we obtain D_{zz} from mean-squared displacement and measure the force autocorrelation function directly, we can obtain the effective temperature:

$$k_B T^{\text{eff}} = \frac{2 D_{zz} k_B T}{D_0 + D_{zz} - (\beta D_0)^2 \int_0^\infty dt \, \langle F_{1z} \exp(\Omega t) F_{1z} \rangle}. \tag{14}$$

Eq. (14) shows that it is possible to monitor FDT violation using Brownian Dynamics simulations of the stationary, unperturbed state.

Before turning to the derivation of MCT expressions we first re-write memory function expression (7). We define an irreducible evolution operator Ω^{irr},

$$\Omega^{\mathrm{irr}} = -\hat{Q}_s \sum_l \frac{\partial}{\partial \mathbf{r}_l} \hat{Q}_s \cdot \left(-\frac{\partial}{\partial \mathbf{r}_l} + \beta \mathbf{F}_l + \mathbf{v}(\mathbf{r}_l) \right) \hat{Q}_s, \quad (15)$$

and then we use standard projection operator manipulations to obtain the following identity:

$$D_0 \delta_{\mathbf{k}_1,\mathbf{k}_3} - \left\langle \mathbf{j}_s(\mathbf{k}_1) \frac{1}{z - \hat{Q}_s \Omega \hat{Q}_s} \left(2\mathbf{j}_s^{\mathrm{eff}}(-\mathbf{k}_3) - \mathbf{j}_s(-\mathbf{k}_3) \right) \right\rangle = \sum_{\mathbf{k}_4} \left(\xi_0 \delta_{\mathbf{k}_1,\mathbf{k}_4} + \beta \xi_0^2 \left\langle \mathbf{j}_s(\mathbf{k}_1) \frac{1}{z - \Omega^{\mathrm{irr}}} \mathbf{j}_s^{\mathrm{eff}}(-\mathbf{k}_4) \right\rangle \right)^{-1} \times \left(k_B T \delta_{\mathbf{k}_4,\mathbf{k}_3} - \xi_0 \left\langle \mathbf{j}_s(\mathbf{k}_4) \frac{1}{z - \Omega^{\mathrm{irr}}} \left(\mathbf{j}_s^{\mathrm{eff}}(-\mathbf{k}_3) - \mathbf{j}_s(-\mathbf{k}_3) \right) \right\rangle \right). \quad (16)$$

Note that here $(...)^{-1}$ denotes the kernel of the inverse integral operator; also, ξ_0 is the friction coefficient of an isolated colloidal particle, $\xi_0 = 1/\mu_0$.

Identity (16) allows us to define the tagged particle friction tensor (the inverse of the first factor at the RHS of Eq. (16)) and an effective temperature (the second factor at the RHS of Eq. (16)). It can be shown that in the long-time, small wavevector limit, the former reduces to the inverse of the mobility tensor, Eq. (13).

To derive MCT expressions for the friction tensor and the effective temperature we follow the standard procedure [19, 20]. We project the currents on the part of the joint density of the tagged particle and of other particles that is orthogonal to the tagged particle density:

$$\mathbf{j}_s(-\mathbf{k}) = \sum_{\mathbf{k}_1,\ldots,\mathbf{k}_4} n_2(-\mathbf{k}_1,-\mathbf{k}_2) g(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4) \times \langle n_2(\mathbf{k}_3,\mathbf{k}_4) \mathbf{j}_s(-\mathbf{k}) \rangle, \quad (17)$$

$$\mathbf{j}_s^{\mathrm{eff}}(-\mathbf{k}) \approx \sum_{\mathbf{k}_1,\ldots,\mathbf{k}_4} n_2(-\mathbf{k}_1,-\mathbf{k}_2) g(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4) \times \langle n_2(\mathbf{k}_3,\mathbf{k}_4) \mathbf{j}_s^{\mathrm{eff}}(-\mathbf{k}) \rangle. \quad (18)$$

In Eqs. (17-18) $n_2(\mathbf{k}_1,\mathbf{k}_2)$ is the part of the joint density of the tagged particle and of other particles that is orthogonal to the tagged particle density, $n_2(\mathbf{k}_1,\mathbf{k}_2) = \hat{Q}_s \sum_{l>1} e^{-i\mathbf{k}_1\cdot\mathbf{r}_1 - i\mathbf{k}_2\cdot\mathbf{r}_l}$, and $g(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4)$ is the inverse of $\langle n_2(\mathbf{k}_3,\mathbf{k}_4) n_2(-\mathbf{k}_5,-\mathbf{k}_6)\rangle$. We use factorization approximation for g, $g(\mathbf{k}_1,\mathbf{k}_2;\mathbf{k}_3,\mathbf{k}_4) \approx \delta_{\mathbf{k}_1,\mathbf{k}_3}\delta_{\mathbf{k}_2,\mathbf{k}_4} (NS(\mathbf{k}_2))^{-1}$, where $S(\mathbf{k}_2)$ is the stationary, shear-rate dependent structure factor. One should note that Eq. (17) is exact for pairwise-additive interactions whereas Eq. (18) constitutes an approximation.

The average in Eq. (17) can be expressed in terms of the direct correlation *force* $\mathbf{C}(\mathbf{k})$ [21],

$$\langle n_2(\mathbf{k}_1,\mathbf{k}_2) \mathbf{j}_s(-\mathbf{k}) \rangle = -i \delta_{\mathbf{k}-\mathbf{k}_1,\mathbf{k}_2} n D_0 \beta \mathbf{C}(\mathbf{k}_2) S(\mathbf{k}_2), \quad (19)$$

(here n is the number density, $n = N/V$) whereas the average in Eq. (18) can be expressed in terms the non-equilibrium, shear-rate-dependent direct correlation function $c(\mathbf{k}) = (S(\mathbf{k}) - 1)/(nS(\mathbf{k}))$,

$$\langle n_2(\mathbf{k}_1,\mathbf{k}_2) \mathbf{j}_s^{\mathrm{eff}}(-\mathbf{k}) \rangle = -i\mathbf{k}_2 \delta_{\mathbf{k}-\mathbf{k}_1,\mathbf{k}_2} n D_0 c(\mathbf{k}_2) S(\mathbf{k}_2). \quad (20)$$

Combining Eqs. (17-20) with Eq. (16) we can obtain expressions for the interaction contributions to the friction tensor and the effective temperature in terms of integrals involving a four-particle, time-dependent correlation function. We factorize this function in terms of the self-intermediate scattering function $F_s(\mathbf{k}_1;\mathbf{k}_2;t)$ and a collective intermediate scattering function $F(\mathbf{k}_1;\mathbf{k}_2;t)$,

$$F(\mathbf{k}_1;\mathbf{k}_2;t) = \frac{1}{N} \langle n(\mathbf{k}_1) \exp(\Omega t) n(-\mathbf{k}_2) \rangle, \quad (21)$$

where $n(\mathbf{k}_1)$ is the Fourier transform of the microscopic density, $n(\mathbf{k}_1) = \sum_l e^{-i\mathbf{k}_1\cdot\mathbf{r}_l}$. As a result we obtain the following mode-coupling expressions for the interaction contributions to the friction tensor and the effective temperature in the long-time (*i.e.* $z \to 0$) limit:

$$-\beta\xi_0^2 \left\langle \mathbf{j}_s(\mathbf{k}_1) \left(\Omega^{\mathrm{irr}}\right)^{-1} \mathbf{j}_s^{\mathrm{eff}}(-\mathbf{k}_2) \right\rangle \approx \frac{n}{V} \sum_{\mathbf{k}_3,\ldots,\mathbf{k}_6} \int_0^\infty dt\; \delta_{\mathbf{k}_1-\mathbf{k}_3,\mathbf{k}_4} \mathbf{C}(\mathbf{k}_4) F_s(\mathbf{k}_3;\mathbf{k}_5;t) F(\mathbf{k}_4;\mathbf{k}_6;t) c(\mathbf{k}_6) \mathbf{k}_6 \delta_{\mathbf{k}_2-\mathbf{k}_5,\mathbf{k}_6}, \quad (22)$$

$$\xi_0 \left\langle \mathbf{j}_s(\mathbf{k}_1) \left(\Omega^{\mathrm{irr}}\right)^{-1} \left(\mathbf{j}_s^{\mathrm{eff}}(-\mathbf{k}_2) - \mathbf{j}_1(-\mathbf{k}_2) \right) \right\rangle \approx -\frac{nD_0}{V} \sum_{\mathbf{k}_3,\ldots,\mathbf{k}_6} \int_0^\infty dt\; \delta_{\mathbf{k}_1-\mathbf{k}_3,\mathbf{k}_4} \mathbf{C}(\mathbf{k}_4) F_s(\mathbf{k}_3;\mathbf{k}_5;t) F(\mathbf{k}_4;\mathbf{k}_6;t)$$

$$\times \left(c(\mathbf{k}_6)\mathbf{k}_6 - \beta \mathbf{C}(\mathbf{k}_6)\right) \delta_{\mathbf{k}_2-\mathbf{k}_5,\mathbf{k}_6}. \tag{23}$$

Expression (22) differs from one derived before [14]: one of the vertices in Eq. (22) involves the direct correlation force, whereas the other involves the non-equilibrium direct correlation function. In the expression obtained in Ref. [14] both vertices were identical and given by the equilibrium direct correlation function. One should note that in equilibrium $\mathbf{C}^{eq}(\mathbf{k}) = k_B T \mathbf{k} c^{eq}(k)$; hence the previous work implicitly used equilibrium approximation for the vertices.

Expression (23) shows that FDT violation is associated with the non-equilibrium character of the stationary, sheared state. In particular, if equilibrium approximation for the vertices is used, no FDT violation is obtained.

One of the most interesting simulational findings is that below the MCT transition temperature FDT violation persists in the limit of the vanishing shear rate [5]. This can be qualitatively understood on the basis of Eq. (23): in the $\dot{\gamma} \to 0$ limit the second vertex in (23) vanishes; however, in the same limit characteristic relaxation times of the self and collective intermediate scattering functions diverge. Thus, it is possible that the expression (23) reaches a finite limit as $\dot{\gamma} \to 0$. In order to prove that this indeed happens one needs to calculate the second vertex in Eq. (23). To this end, it might be possible to use an approach proposed by Fuchs and Cates [10]: using mode-coupling theory to calculate steady state properties by starting from the equilibrium state and considering transient dynamics.

Another very interesting result of Ref. [5] is that in the $\dot{\gamma} \to 0$ limit the same effective temperature is obtained for different wavevectors and even for different observables. Expression (23), in principle, allows one to verify this fact theoretically. In particular one could check whether *tensorial* quantity (23) reduces to a scalar one in the $\dot{\gamma} \to 0$ limit. Again, in order to investigate this, the second vertex is needed.

To summarize, we have derived Green-Kubo-like formulae for the self-diffusion and mobility coefficients, and mode-coupling expressions for the friction tensor and the effective temperature. The numerical analysis of these expressions is left for future work.

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